Experimental report

13/02/2019

Proposal: 7-02-154 **Council:** 10/2014

Title: Freezing of polar nanoregions in PMN single crystal

Research area: Physics

This proposal is a new proposal

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Samples: Pb(Mg1/3,Nb2/3)O3

Instrument	Requested days	Allocated days	From	To
IN16B	7	7	10/07/2015	17/07/2015

Abstract:

We intend to study a Vogel-Fulcher relaxation in the relaxor ferroelectric Pb(Mg1/3,Nb2/3)O3 single crystal via investigation of its diffuse scattering. This study will give valuable information about time-space correlation lengths of polar fluctuations associated with exceptional dielectric properties of multiaxial relaxor perovskites. In this challenging study, we would like to take an advantage of fixed window measurements on the new high-flux IN16B backscattering spectrometer and our experience with this type of experiment.

Freezing of polar nanoregions in a PMN single crystal

The experiment 7-02-154 was performed on the IN16B backscattering instrument on July 10-17, 2015. The aim of this experiment was to study a Vogel-Fulcher relaxation in the relaxor ferroelectric $PbMg_{1/3}Nb_{2/3}O_3$ (PMN) single crystal. This was done via investigation of its diffuse scattering in order to obtain valuable information about time-space correlation lengths of polar fluctuations associated with the Vogel-Fulcher relaxation.

During the experiment, however, there were indications that the sample that we had obtained through collaboration contained also some amount of titanium. Later analysis showed its concertation to be: $Pb(Mg_{1/3}Nb_{2/3})_{0.84}Ti_{0.16}O_3$ (PMN-16%PT), which is also relaxor ferroelectric. The crystal (volume of 4.5 cm^3) was put inside a Nb holder and its crystallographic orientation was rechecked on IN3. Then, the sample was placed in a standard orange cryofurnace and measured in the (*hk0*) scattering plane on IN16B. The IN16B instrument was operated in its standard configuration: an unpolished Si(111) Doppler monochromator, unpolished Si(111) analysers (neutron wavevector of k_f = 1 $Å^{-1}$) and PSD detectors. The instrumental energy resolution was 1 μeV. Most of the measurements were carried out in the 100 Brillouin zone as follows:

1. Small elastic maps around the 100 Bragg reflection were measured at 0 μeV and 3 μeV from 100 to 450 K. The point of this stage was to find the best **Q** at which the diffuse scattering is free of contamination from the 100 Bragg reflection and shows a characteristic temperature change [1].
2. Then, the diffuse scattering at selected momentum transfer **Q** ~ (1.05,0.05,0) was measured at several energy transfers from 550 K to 100 K. The collected data are plotted in Fig. 1. The acquisition times were up to 1 h per point for the highest energy transfer.
3. In the dynamic operation mode, the whole spectra at the same momentum transfer **Q** ~ (1.05,0.05,0) were measured from –6 to 6 μeV at two temperatures (see Fig. 2). The acquisition time was 8 h at 347 K and 3 h at 543 K.

The diffuse scattering in Fig. 1 shows a peak at 300-350 K which is related to the phase transition from cubic to rhombohedral phase [2]. Moreover, the peak shifts to higher temperatures with increasing energy transfer which is a clear signature of the dielectric Vogel-Fulcher-like relaxation – polar fluctuations – observed in relaxor ferroelectrics [2,3]. The inelastic component is also seen from the spectrum at 347 K in Fig. 2.

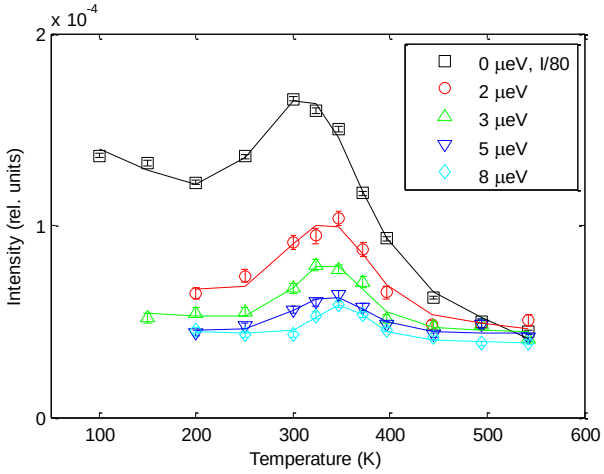

Fig. 1: Temperature dependence of diffuse scattering at **Q** ~ (1.05,0.05,0).

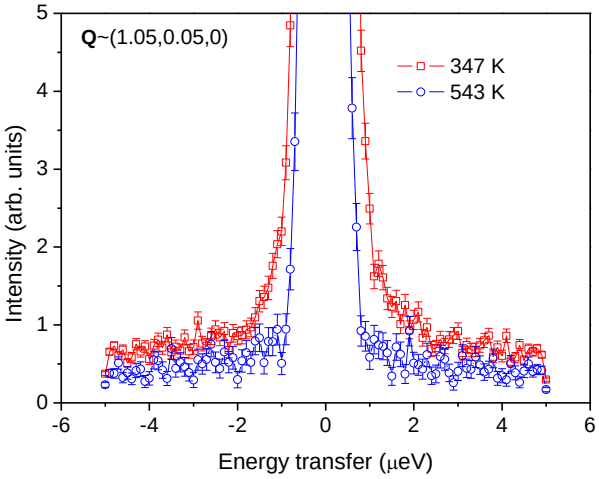

Fig. 2: Inelastic neutron spectra at **Q** ~ (1.05,0.05,0).

In conclusion, measurements at constant energy transfer seem to be extremely advantageous for this type of dynamics in relaxor ferroelectrics. We have found that diffuse scattering in the (100) Brillouin zone corresponds to polar fluctuations seen by dielectric spectroscopy [2] as we proved for the first time in the uniaxial relaxor ferroelectric $Sr_{0.61}Ba_{0.39}Nb_2O_6$ [3].

References:

[1] C. Stock, *et al.*, *Phys. Rev. B* **81**, 144127 (2010).
[2] H. Ohwa *et al.*, *J. Phys. Soc. Jpn.* **70**, 3149 (2001).
[3] P. Ondrejkovic *et al.*, *Phys. Rev. Lett.* **113**, 167601 (2014).